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ABSTRACT

An isomeric state of stable Ba^{137} which decays by γ emission to the ground state with a half-life of 2.5 minutes has been discovered. This metastable state is formed by β^- decay of Cs^{137}. The Ba^{137} γ has an energy of 0.75 Mev and is 13% internally converted giving rise to $\sim$0.7 Mev conversion electrons and K x-rays.

INTRODUCTION

Cs^{137} is listed in the tables[1] as having two betas of 0.5 Mev and 0.8 Mev in approximately equal numbers. In addition, a γ of 0.7–0.75 Mev energy is listed.

β γ coincidence studies[2] showed, however, that the γ was not coincident with either of the beta groups. Two possibilities remained: either the γ was coincident with a very weak beta which was largely absorbed by the counter window or the lifetime of the Ba^{137} nucleus was long compared with the resolution time of the coincidence circuit. The latter possibility was investigated by means of delayed coincidence techniques similar to those used by De Benedetti.[3] No delayed coincidences were found. With the maximum resolving time used (576 μ sec) it was estimated that a metastable state with a half-life of 0.01 second or less would have been detected.

Rapid chemical separations of barium from Cs^{137} were then carried out to look for a metastable state with a half-life greater than a few seconds. The barium fractions showed γ activity which decayed with a half-life of 2.5 minutes.

A detailed account of the experiments which were carried out to establish the decay scheme of Cs^{137} is given in the following sections.

EXPERIMENTAL

Half-life Determination

Cs^{137} was isolated from uranium which had been irradiated with pile neutrons. Two years had elapsed since the end of the irradiation so that all shorter-lived cesium isotopes had decayed. The standard procedure[4] of $CsClO_4$ precipitation from absolute ethyl alcohol followed by $La(OH)_3$ scavenging was followed. Three cycles were carried out.

In one experiment a rapid separation of barium from the cesium tracer solution was carried out by precipitation of $Ba(NO_3)_2$ with fuming nitric acid. The barium nitrate was filtered by suction on to an asbestos mat in a Gooch crucible. The filtration was complete within five seconds after the start of the separation. An Eck and Krebs γ counter was positioned beside the crucible and was connected to a calibrated logarithmic counting rate meter equipped with an Esterline Angus recorder. A half-life

of 2.57 minutes was obtained (Figure 1-A). A background of 1650 c/m, due primarily to the proximity of the filtrate, was subtracted.

In another experiment the barium was separated from cesium by a barium nitrate precipitation and further purified by precipitation as $BaCl_2$. The barium chloride was collected on a filter disc and mounted on a mounting card under 2.5 mg/cm^2 of cellophane. Counting was done on an end window Geiger counter connected to a scaler. The counter was operated continuously and the recorder read at one minute intervals. Counting was begun 3.3 minutes after the beginning of the separation. After subtraction of a 40 c/m background, the decay curve shown in Figure 1-B was obtained. The barium decayed exponentially for ten half-lives with a half-life of 2.53 minutes. An aluminum absorber 186 mg/cm^2 thick was used to reduce the initial counting rate to a reasonable value. The data have been corrected for resolving time losses by means of a linear correction of 0.3% per 1000 c/m.

In order to verify the cesium-barium parent-daughter relationship, a rapid separation of cesium was performed and the growth of the γ activity of the barium observed. The cesium was precipitated as $CsClO_4$ with 70% $HClO_4$ and absolute ethyl alcohol and filtered on a Gooch crucible. The γ counter was placed next to the crucible and the counting rate recorded on an Esterline Angus recorder as in the first barium separation. The growth curve of barium γ activity is shown in Figure 2. Subtraction of the observed counting rates from the saturation activity gives a half-life of 2.49 minutes for the barium. The extrapolation of this subtracted curve to the time of cesium separation gives a counting rate which is within 3% of the saturation γ counting rate. This indicates that, within experimental error, all of the γ activity arises from the 2.5 minute metastable state.

Absorption Curves

Aluminum and lead absorption curves were taken of the radiations emitted by separated barium. The barium was precipitated first as the nitrate and then as the chloride. This procedure was found to give a decontamination factor of 50,000 from cesium. The absorption curves were started with the heaviest absorbers and progressively lighter absorbers used as the sample decayed. In this way counting rates between 2000 and 10,000 c/m were obtained for each absorption point.

The aluminum absorption curve was taken on the first shelf position of an end window counter having a mica window 3.2 mg/cm^2 thick. This curve (Figure 3) shows a γ component and a beta component. The beta component shows the flattening near zero absorber which is characteristic of monoenergetic electrons and may be ascribed to conversion electrons produced by internal conversion of the barium γ. The visual range is 300 mg/cm^2, corresponding to an electron energy of $\sim$ 0.76 Mev.

The lead absorption curve (Figure 4) for separated barium was taken on the second shelf of the end window counter used for the aluminum absorption curve. An aluminum absorber of thickness 342 mg/cm^2 was placed over the sample to absorb the beta rays. A single component of half-thickness 7.4 g/cm^2 is obtained corresponding to a γ energy of 0.74 Mev. This agrees well with the value of 0.7 – 0.75 Mev listed for Cs^{137}.

A copper absorption curve was taken of barium in equilibrium with Cs^{137} in a search for x-rays accompanying internal conversion of the barium γ. An Eck and Krebs tube filled with Kr instead of the usual argon filling was used in order to increase the counting efficiency for x-rays. The absorption curve (Figure 5) may be broken into two components, one with a half-thickness of 10.2 g/cm^2 (0.7 Mev) and another with a half-thickness of 51 mg/cm^2 (28 Kev). The harder component is the unconverted γ whereas the softer component has an energy close to that expected for barium K x-rays (32 Kev for Kα).

Coincidence Measurements

$\beta\,\gamma$ coincidence measurements were carried out as a check on the preceding work. If the barium γ is converted internally, coincidences between conversion electrons and x-rays may be expected.

The coincidence circuit was a modification of one described by Bradley and Epstein[5] and had a

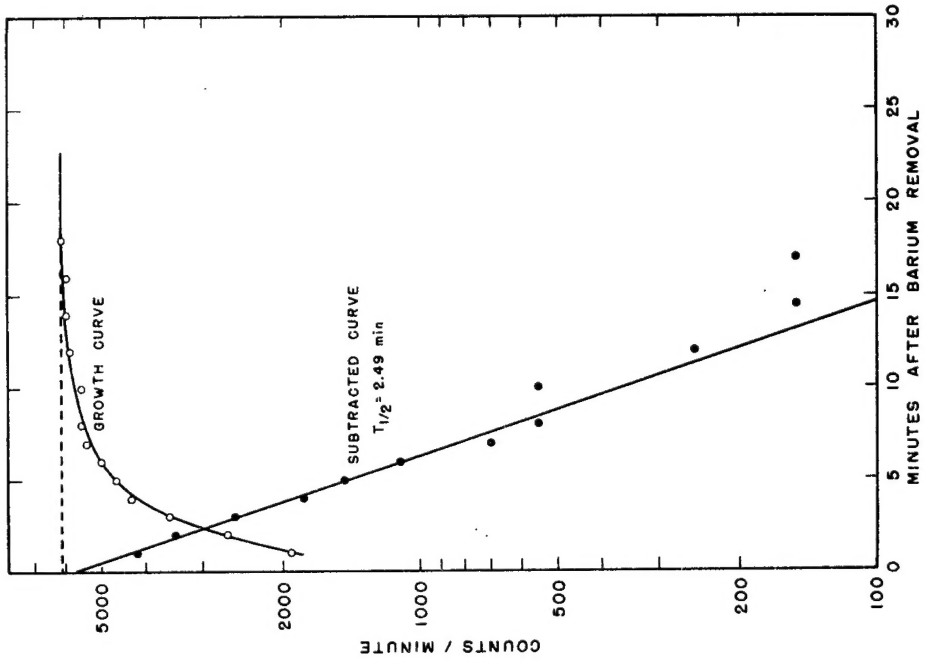

Figure 2. Growth of Ba^{137} γ activity in Cs^{137}.

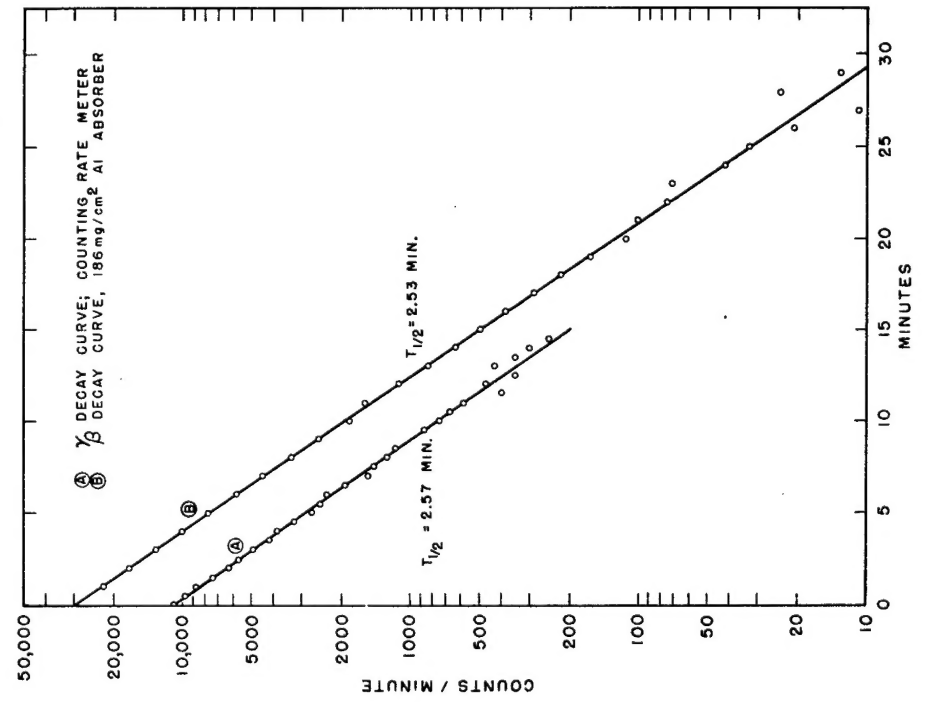

Figure 1. Decay curves of Ba^{137} separated from Cs^{137}.

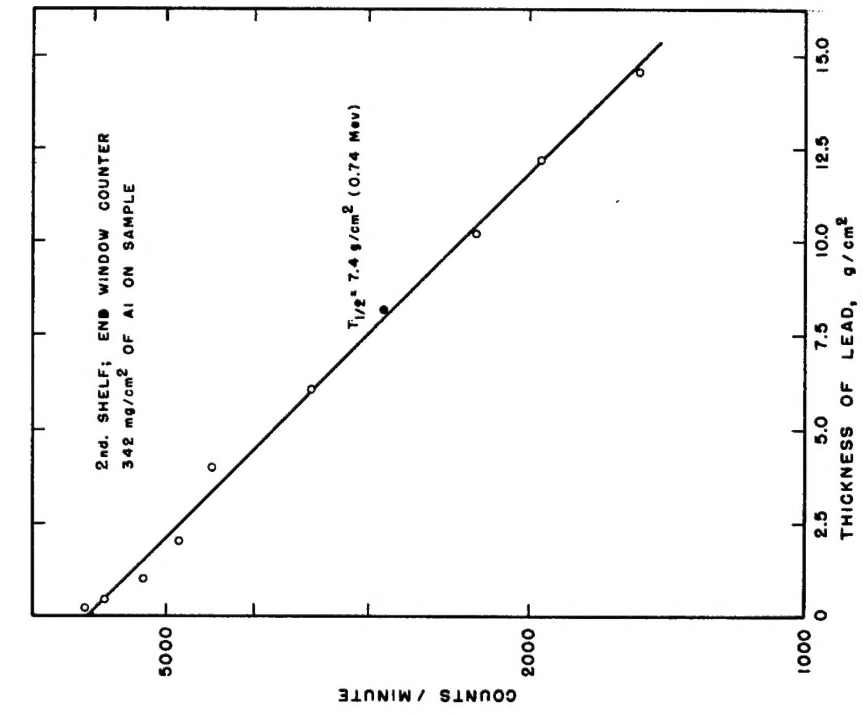

Figure 4. Lead absorption curve of Ba^{137}.

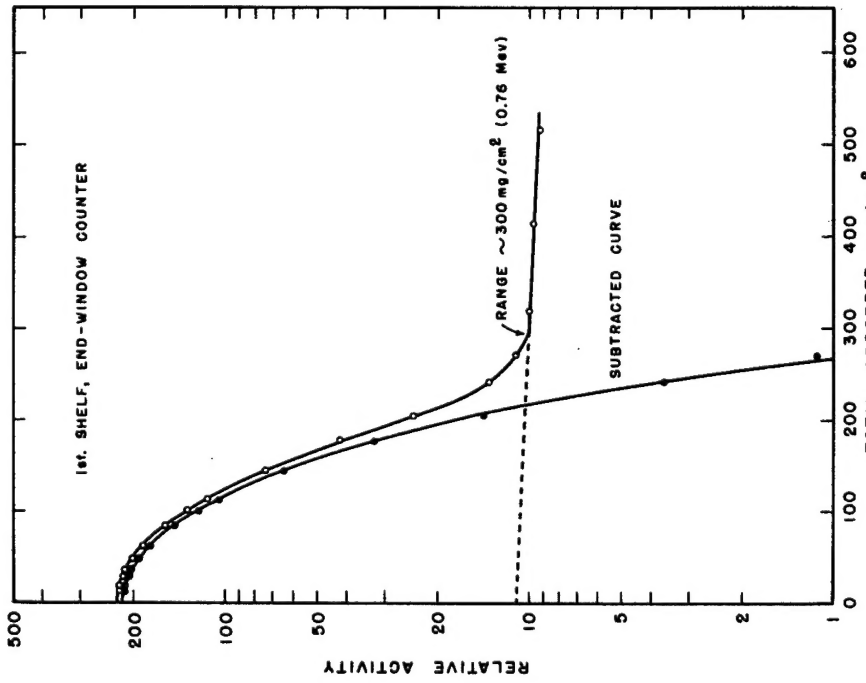

Figure 3. Aluminum absorption curve of Ba^{137}.

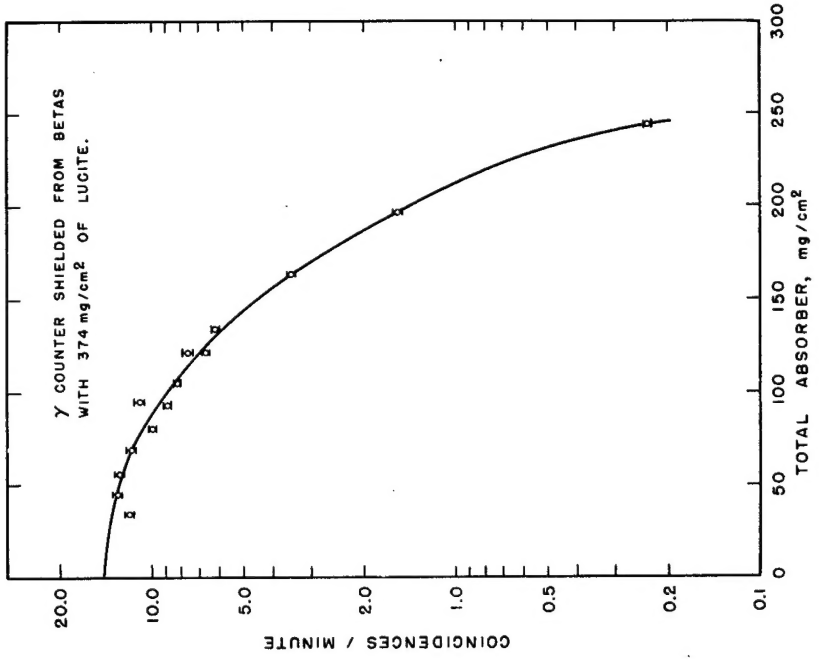

Figure 6. Al β absorption curve of $\beta\,\gamma$ coincidences in Ba^{137}.

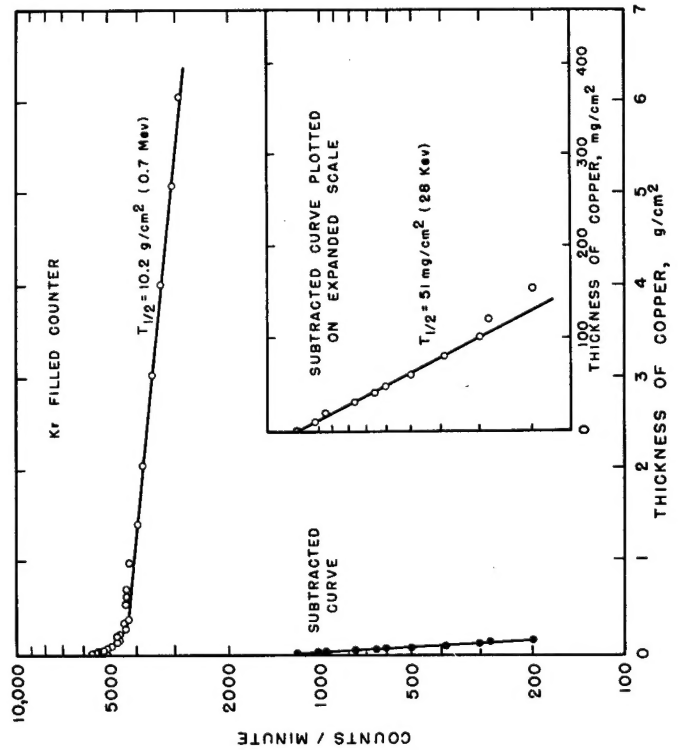

Figure 5. Copper absorption curve of Ba^{137} in equilibrium with Cs^{137}.

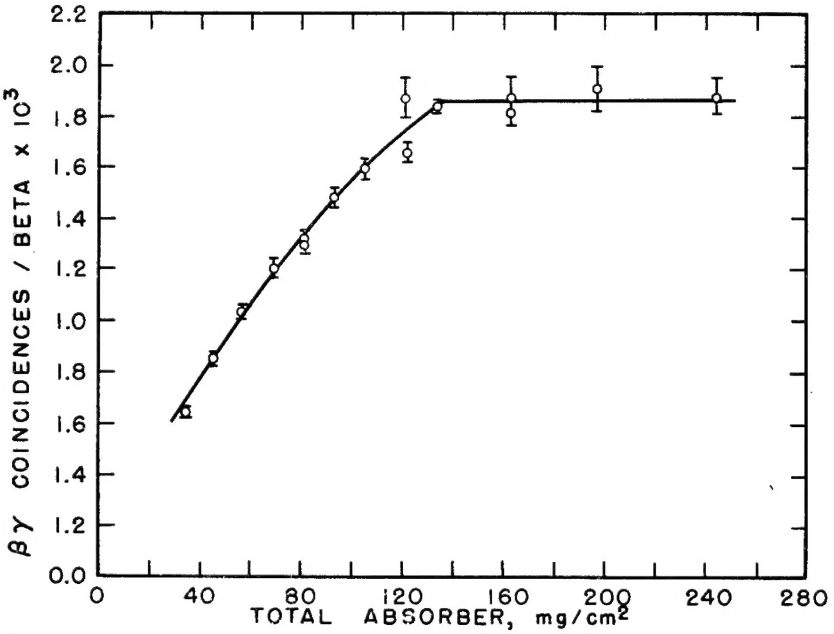

Figure 7. X-ray-conversion electron coincidences in Cs^{137}-Ba^{137} aluminum absorption of betas.

resolving time of approximately one microsecond. Two thin-walled Eck and Krebs tubes were placed in a large copper lined box which provided electrostatic shielding for the counters. The box was shielded on the sides and bottom with four inches of lead. A 1/4 inch thick aluminum shield was placed between the counters to isolate the counters electrically and to reduce scattering of beta particles from one counter to the other. The aluminum shield had an opening cut in it to accommodate the sample card. The beta counter was filled to a pressure of 9 cm of argon and 1 cm of ethyl alcohol while the γ counter was filled to 9 cm of krypton and 1 cm of amyl acetate. It was found experimentally that the use of a krypton filling increased the efficiency of the γ counter for barium K x-rays by a factor of four over that obtained with argon. The counters were aligned with their axes parallel and 3.9 cm apart with the sample centered between them. In all these experiments a Lucite absorber 374 mg/cm^2 thick was placed between the sample and the γ counter. The observed coincidence rates were corrected for chance and cosmic ray coincidences.

An aluminum beta absorption curve of the $\beta\ \gamma$ coincidences in an equilibrium cesium-barium sample was obtained by placing sheets of aluminum between the sample and the beta counter (Figure 6). Two samples, one five times as strong as the other, were used to cover the full range of absorbers. The curve is nearly identical with the absorption curve for the Ba^{137} conversion electrons alone (Figure 3). A plot of the coincidence rate per recorded beta particle is shown in Figure 7. It may be seen that $\beta\ \gamma/\beta$ rises from a value of 0.6×10^{-3} at 30 mg/cm^2 of absorber to a value of 1.86×10^{-3} at 140 mg/cm^2. The value of $\beta\ \gamma/\beta$ then remains constant out to the highest absorber used, 244 mg/cm^2. These results are in accord with the postulate that the coincidences are between the conversion electrons and the x-rays of barium. The cesium disintegration betas and the unconverted, delayed γ cannot be coincident with each other nor with the conversion electrons or x-rays. The increase in $\beta\ \gamma/\beta$ with increasing absorber is due to absorption of the cesium betas; after essentially all of the cesium betas have been absorbed, $\beta\ \gamma/\beta$ remains constant.

A copper γ absorption curve of the $\beta\ \gamma$ coincidences was also taken (Figure 8). The γ counter was shielded from betas by 374 mg/cm^2 of Lucite. An aluminum absorber 41 mg/cm^2 thick was placed between the sample and the beta counter. This absorber, added to the $\sim$30 mg/cm^2 window of the beta counter, served to filter out most of the cesium betas and thus permitted the use of a more active

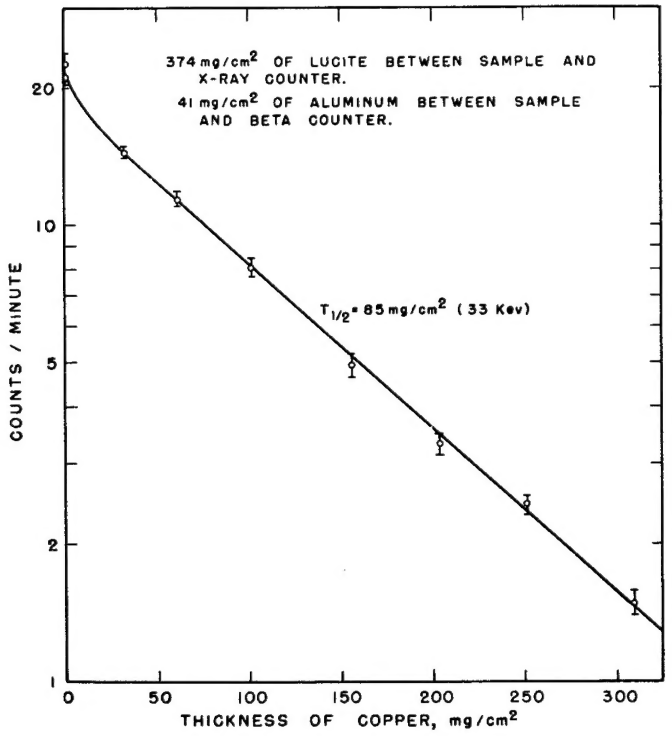

Figure 8. Copper x-ray absorption curve of x-ray-conversion electron coincidences in Ba^{137}.

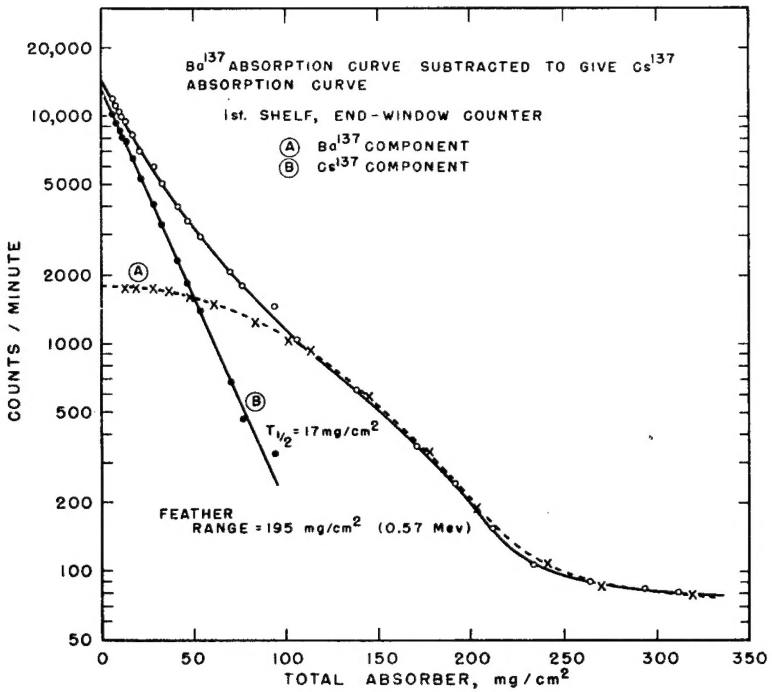

Figure 9. Aluminum absorption curve of Cs^{137}-Ba^{137} in equilibrium.

sample than would otherwise have been possible. The main part of the absorption curve shows an 85 mg/cm^2 half-thickness (33 Kev) which corresponds to that expected for the K x-rays of barium (K_α = 32 Kev). The slight curvature at the beginning may be due to L x-rays.

Conversion Coefficient and Probable Decay Scheme

Since the radiations from Ba^{137} are now fairly well established as consisting of an $\sim$0.75 Mev γ, conversion electrons from this γ, and K x-rays, it is therefore possible to obtain more information about the radiations from Cs^{137} by means of absorption curves taken on an equilibrium barium-cesium sample.

An aluminum absorption curve of Cs^{137} in equilibrium with Ba^{137} is shown in Figure 9. This curve was taken under the same conditions as the aluminum absorption curve of the separated barium. In addition to the barium γ background, this curve shows only two components; the $\sim$ 0.75 Mev conversion electron component (A) and the softer beta component from the decay of the cesium. Subtraction of the conversion electron and γ components from the total curve gives the beta absorption curve of cesium (B). Feather analysis gives a maximum beta energy of 0.57 Mev. A careful search was made for higher energy betas which might arise from beta transitions directly to the ground state of Ba^{137}. None were found. It was estimated that 2% branching to give a 1.3 Mev beta could have been detected. It appears from this that Cs^{137} has a simple beta spectrum. If this is correct, the conversion coefficient of the barium γ may be calculated by comparison of the extrapolated counting rates of the barium conversion electrons and the disintegration betas of the cesium. An analysis of the aluminum absorption curve of Figure 9 gives a conversion coefficient of 0.13.

On the basis of the preceding data, a decay scheme is proposed in Figure 10.

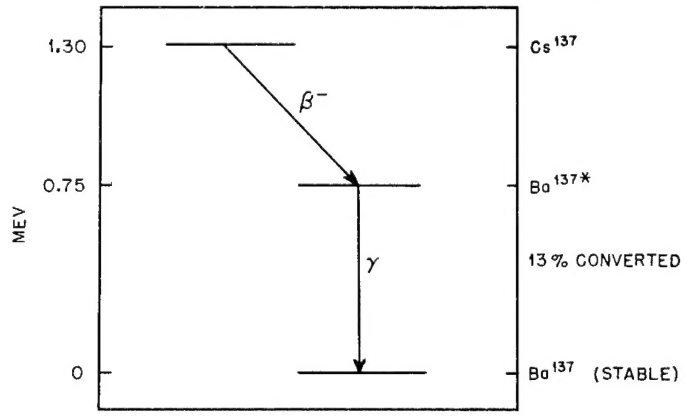

Figure 10.

A point of further interest is that assignment of the hard beta component in the equilibrium cesium-barium aluminum absorption curve to barium will cause a change in the accepted value for the half-life of Cs^{137}. Cs^{137} has been assigned a half-life of 33 $\pm$ 3 years[1] on the basis of its observed disintegration rate and estimated fission yield in U^{235}. Lowering the observed counting rate by 13% to give the counting rate of Cs^{137} alone will increase the calculated half-life to 37 $\pm$ 3 years.

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5. Bradley, W. H. and B. Epstein, CP-2735, February, 1945.